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REVIEW OF LITERATURE ON WASTE SOLIDIFICATION/STABILIZATION WITH EMPHASIS ON METAL-BEARING WASTES

ABSTRACT   Heavy metal contamination is the second most common contaminant as past Naval disposal sites. Various solidification/stabilization processes are available to meake wast non-hazardous. Their effectiveness depends on the type, combination, and concentration of contaminants in the waste.

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NAVAL CIVIL ENGINEERING LABORATORY   PORT HUENEME   CALIFORNIA 93043

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METRIC CONVERSION FACTORS

Approximate Conversions to Metric Measures

Symbol	When You Know	Multiply by	To Find	Symbol
		LENGTH		
in	inches	*2.5	centimeters	cm
ft	feet	30	centimeters	cm
yd	yards	0.9	meters	m
mi	miles	1.6	kilometers	km
		AREA		
in^2	square inches	6.5	square centimeters	cm^2
ft^2	square feet	0.09	square meters	m^2
yd^2	square yards	0.8	square meters	m^2
mi^2	square miles	2.6	square kilometers	km^2
	acres	0.4	hectares	ha
		MASS (weight)		
oz	ounces	28	grams	g
lb	pounds	0.45	kilograms	kg
	short tons (2,000 lb)	0.9	tonnes	t
		VOLUME		
tsp	teaspoons	5	milliliters	ml
Tbsp	tablespoons	15	milliliters	ml
fl oz	fluid ounces	30	milliliters	ml
c	cups	0.24	liters	l
pt	pints	0.47	liters	l
qt	quarts	0.95	liters	l
gal	gallons	3.8	liters	l
ft^3	cubic feet	0.03	cubic meters	m^3
yd^3	cubic yards	0.76	cubic meters	m^3
		TEMPERATURE (exact)		
°F	Fahrenheit temperature	5/9 (after subtracting 32)	Celsius temperature	°C

*1 in = 2.54 (exactly). For other exact conversions and more detailed tables, see NBS Misc. Publ. 286, Units of Weights and Measures, Price $2.25, SD Catalog No. C13.10:286.

Approximate Conversions from Metric Measures

Symbol	When You Know	Multiply by	To Find	Symbol
		LENGTH		
mm	millimeters	0.04	inches	in
cm	centimeters	0.4	inches	in
m	meters	3.3	feet	ft
m	meters	1.1	yards	yd
km	kilometers	0.6	miles	mi
		AREA		
cm^2	square centimeters	0.16	square inches	in^2
m^2	square meters	1.2	square yards	yd^2
km^2	square kilometers	0.4	square miles	mi^2
ha	hectares (10,000 m^2)	2.5	acres	
		MASS (weight)		
g	grams	0.035	ounces	oz
kg	kilograms	2.2	pounds	lb
t	tonnes (1,000 kg)	1.1	short tons	
		VOLUME		
ml	milliliters	0.03	fluid ounces	fl oz
l	liters	2.1	pints	pt
l	liters	1.06	quarts	qt
l	liters	0.26	gallons	gal
m^3	cubic meters	35	cubic feet	ft^3
m^3	cubic meters	1.3	cubic yards	yd^3
		TEMPERATURE (exact)		
°C	Celsius temperature	9/5 (then add 32)	Fahrenheit temperature	°F
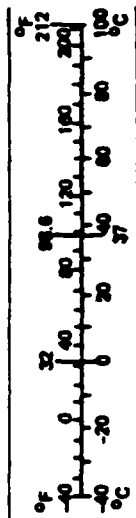
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Heavy metal contamination is the second most common contaminant at past Naval disposal sites. Various solidification/stabilization processes are available to make waste non-hazardous. Their effectiveness depends on the type, combination, and concentration of contaminants in the waste.

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EXECUTIVE SUMMARY

by

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This report provides a starting point for the Navy's RDT&E program on the solidification and stabilization of heavy metal bearing wastes. Many vendors of this technology have recently entered the market place with their often proprietary processes. Because the vendors are reluctant to identify the chemical used in their process, much confusion exists as to the available types and capabilities of the processes.

This report describes the types of available processes, identifies vendors of these processes, explains the chemistry behind two commonly used processes, and discusses factors affecting the long term stability of the treated waste. A vendors' reference list, cost comparison table, and waste compatibility table are included to help the reader find more information on solidification/ stabilizations processes.

Solidification and stabilization of heavy metal bearing wastes promises to be effective in converting hazardous wastes into non-hazardous materials. Solidification, a process in which materials are added to a waste to produce a solid object, and stabilization, a process by which a waste is converted to a more chemically stable form, are synonymous with the term chemical fixation. Four commonly available types of processes are cement-based, pozzolanic, thermoplastic, and organic polymer-based. Two of these processes, cement-based and pozzolanic, are most extensively used on heavy-metal containing wastes.

The silicate process uses soluble silicates and cement to physically encapsulate the particles in the waste and form a protective coating. Typically, the silicate and cement are added in concentrations sufficient to create a blocky texture but not complete solidification.

The sulfide process uses sodium sulfide and cement to chemically react with the waste and form highly insoluble sulfide salts of heavy metals. Certain heavy metals such as chromium, selenium, and arsenic do not form insoluble sulfide salts and are poorly suited to fixation by this process. Heavy metals that do form a highly insoluble sulfide salt may be better fixated by this process than the silicate process. Care must be taken to ensure that the treated waste does not exceed the RCRA corrosivity guideline of pH 12.5 and the RCRA reactive sulfides guideline of 500 parts per million. The physical form of the waste changes relatively little during stabilization.

As vendors' claims routinely exceed the current capabilities of this technology, additional RDT&E is needed to transfer solidification/stabilization processes to routine use. Long term stability issues need to be resolved with regulatory agencies. Also, better screening and cost estimation procedures are needed in order to determine the proper process for each type of waste and to develop a standard statement of work.

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WITH EMPHASIS ON METAL-BEARING WASTES

to

NAVAL CIVIL ENGINEERING LABORATORY

by

Sandra Clark
Talya Greathouse
Jeffrey Means

BATTELLE

INTRODUCTION

Definition of Terms for Solidification/ Stabilization Technologies

Solidification is a process in which materials are added to a waste to produce a solid. Stabilization is a process by which a waste is converted to a more chemically stable form. These two processes are frequently combined and referred to as a single process known as solidification/stabilization (S/S). S/S processes are used to convert hazardous materials into nonhazardous materials that are acceptable for disposal under applicable federal, state, and local standards. The primary goals of S/S processes are:

- To improve handling and physical characteristics of waste
- To prevent leaching of contaminants into the environment
- To detoxify the hazardous constituents of a waste when possible.

The term "chemical fixation" appears frequently in the literature. For the purposes of this report, chemical fixation or some variation of that term is synonymous with solidification/stabilization.

Scope and Approach

Stricter regulations on land disposal of hazardous materials have prompted research and development of improved S/S methods. In addition, S/S technologies have attempted to develop strong, durable, impermeable materials that have potential reuse value (e.g., treated hazardous waste used as construction material). S/S processes differ from most types of waste treatment processes in that waste constituents are not destroyed or converted into other compounds or by-products. The waste constituent remains intact, but its physico-chemical form is changed to inhibit leaching.

This report gives a brief overview of: a) the types of available solidification/stabilization processes; b) the chemicals used in these processes; c) the waste types that these processes can handle; d) the fixation mechanisms; e) the stability of the final waste product; f) the use/reuse potential of the final treated waste product; g) waste stabilization costs; and h) a brief discussion of relevant DoD (other than Navy) activities in this area. Emphasis is placed on technologies for the treatment of hazardous solid wastes containing metals, such as contaminated sludges and soils, for the purpose of reclassifying them as nonhazardous. The treated wastes may then be removed to a sanitary (as opposed to secure) landfill or recycled. S/S processes for organic wastes are mentioned for completeness but are not the principal concern. In situ treatment technologies are briefly mentioned but are also not a major focus. Treatment technologies for liquid wastes were specifically excluded from this review.

The major sources of information were the following:
a) discussions with and information provided by EPA-Cincinnati staff in the Waste Stabilization Section of the Municipal Solid Waste and Residuals Management Branch of the Risk Reduction Engineering Laboratory;
b) information provided by some 20 - 25 vendors of waste stabilization processes, including vendors involved in the EPA Superfund Innovative Technology Evaluation (SITE) program; and c) reports and articles identified in a literature search of a computer-based data base. References

both cited in the text and consulted but not cited are provided at the conclusion in Appendix A.

TYPES OF PROCESSES AVAILABLE

The major types of processes that are commonly available to stabilize, solidify, or fixate waste materials are cement-based, pozzolanic, thermoplastic, and organic polymer-based processes:

- Cement-based - This process relies on one of the Portland cement types to help bind waste constituents into a matrix that improves the properties of the waste. The process, when properly applied, typically improves the handling ability of wastes and in most cases decreases leachable components. Soluble silicates or sodium sulfide may be added to enhance metal containment.

- Pozzolanic (lime-based) - Pozzolan-based processes rely on pozzolanically active materials to bind the constituents of a compatible waste into a matrix which exhibits improved handling and leaching characteristics. Pozzolans are materials that display no cementing action when alone, but when combined with lime and water at ordinary temperatures, form cementitious substances. Examples of common pozzolans are fly ash, pumice, lime kiln dusts, and blast furnace slag. Soluble silicates or sodium sulfide may be added to enhance metal containment.

- Thermoplastic (asphaltic) - These processes rely on the use of a thermoplastic binder, such as asphalt, to bind the waste components into a solidified, impermeable matrix. A thermoplastic is an organic polymer that is fluid at high temperatures, but behaves as a solid at ordinary temperatures.

- Organic Polymer (urea-formaldehyde) - This process (the urea-formaldehyde system is used almost exclusively) relies on polymer formation to bind the waste. The waste and a monomer are first thoroughly mixed, then a catalyst is added which initiates the formation of the polymer.

Cement-based and pozzolanic processes or a combination of both are the methods of choice in the S/S industry today. Approximately 75 percent of the vendors contacted used these processes exclusively. This is likely attributable to the low cost of the cement-pozzolanic processes, their applicability to treating a wide variety of waste types, and the ease with

which they are implemented in the field (Wiles and Apel, undated). Asphaltic processes are also frequently used and are gaining popularity as a means of converting waste having a suitable physical form (e.g., sandblast grit) to a stable reusable product.

More innovative solidification/stabilization processes have also been proposed for the treatment of hazardous wastes. In situ vitrification (ISV), for example, has been suggested as a suitable process for stabilizing soil contaminated with radioactivity. This process involves inserting electrodes around a volume of contaminated soil, passing a current through the soil to produce a molten mass, and eventually forming a final waste product that resembles obsidian or basalt glass (Jacobs, undated). Another innovative process utilizes molten sulfur to stabilize hazardous waste (Bell et al., 1981). Lastly, glass-forming compounds have been used to stabilize nonvolatile inorganics by encapsulating the waste in the final glass product. Four major types of glass are suitable for this process, namely, phosphate glass, borosilicate glass, glass ceramic, and supercalcine (Bell et al., 1981).

There are other processes that are also available or under development, such as sorption, macro-encapsulation (plastic jacketing), sintering, and self-cementing techniques. Sorption is employed fairly frequently and entails using chemicals such as activated carbon, anhydrous sodium silicate, gypsum, or clay to take up free liquid. Although a solid is produced, this process does not necessarily reduce the leaching potential of the contaminant from the treated waste. The other processes mentioned above are not generally widely used, however, because of their high cost or experimental nature.

Solidification/stabilization processes are generally nondestructive and do not remove or reduce the amounts of any of the hazardous constituents in the waste. The concentration of hazardous constituents in the treated waste is typically slightly lower than in the untreated waste, due to dilution by the treatment chemicals; however, the principal effect of S/S is to either physically encapsulate or change the physico-chemical form of the pollutant in the waste, resulting in a less leachable product.

There is frequently some confusion regarding the texture of the stabilized material. The term, solidification/stabilization, connotes hardened products with the texture of bricks. In fact numerous S/S products have a granular texture not too dissimilar from the waste that was stabilized. There is a conscious attempt by the industry to minimize the volume of the product, for at least 2 reasons. One is to conform to the waste minimization requirements of RCRA. The other relates to cost, in that treatment chemicals are expensive and disposal fees are based on volume. Certain wastes can be stabilized with as little as 10 percent additional volume in treatment chemicals, whereas solidification processes typically result in a volume expansion of 100 percent or more.

It should also be noted that, while federal statutes base a hazard classification upon the soluble or leachable concentration of metal in a waste, certain states, such as California, may classify a metal-contaminated waste hazardous by either its total or soluble metal concentrations. While the soluble metal content criteria are usually more restrictive for landfill disposal, it is conceivable that a waste could conform to the soluble thresholds but still be classified as hazardous because it exceeds one or more of the total metal thresholds. In this case, waste stabilization would be of no use in converting the waste to a nonhazardous form.

CHEMICALS USED

Fly ash and cement are commonly used to solidify hazardous waste. Table 1 illustrates the basic chemical differences between the two materials. Both materials are relatively low in cost and easy to use; however, both share the disadvantage of increasing the total volume of end product. In addition to fly ash and cement, other inorganic binders include lime, gypsum, and silicates (Wiles, 1987).

Recently, research has focused on adding agents (called chemisorbents) to inorganic binders. Chemisorbents chemically react with the binders but also provide sites for reacting with the waste contaminant. This process involves incorporation of the adsorbed contaminant into the

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cement matrix rather than entrapment of the contaminant in the voids of the cement matrix; thus, the use of chemisorbents tends to reduce the leaching potential for certain contaminants. Ion exchange resins, clay, and zeolites are several examples of chemisorbents (Wiles, 1987).

Numerous proprietary additives are used by various vendors to enhance specific chemical or physical properties involved in their S/S processes. Soliditech Inc., for instance, uses a reagent known as URRICHEM in their cement-based technique to prevent "flash setting" while enhancing hydration and matrix formation. Sodium sulfide is a common additive for fixing metal-bearing wastes, because it forms insoluble precipitates with a wide variety of heavy metals. Soluble silicates (usually sodium silicate) and lime are also common S/S additives. A study by Johannesmeyer and Ghosh (1984) showed that soluble silicate was more effective than elevated pH in stabilizing chromium and cadmium in electroplating wastewater sludge.

In addition to inorganic binders, numerous organic binders have been used to solidify/stabilize hazardous wastes. Organic binders are more costly and more difficult to use than inorganic binders; yet, a minimal increase in volume of the end product and a significant increase in performance often make organic binders the most appropriate S/S process. Organic binders include epoxy, polyesters, asphalt/bitumen, polyolefin (primarily polyethylene and polyethylene-polybutadiene), and most commonly urea-formaldehyde (Wiles, 1987). Combinations of inorganic and organic binder systems have also been used. These include diatomaceous earth with cement and polystyrene; polyurethane and cement; and polymer gels with silicate and lime cement (Wiles, 1987).

The chemicals used to treat a waste not only need to be effective in waste stabilization, but should also be nontoxic, noncorrosive, and generally compatible with disposal environment. For example, if too much lime or caustic is added during the stabilization process, then the pH of the waste may exceed the RCRA limit of 12.5. Also, if sulfides are used, the U.S. EPA reactive sulfide (as determined in a test where an aliquot of waste is dispersed in a pH 2 acid solution and any H_2S formed is purged and quantitated) guideline of 500 mg/kg may be exceeded. However, it appears

that these potential problems can be avoided in most cases by process modification.

APPLICABLE WASTE TYPES

Table 2 presents a broad overview of waste processes discussed above and their potential compatibility with different waste components. This table shows that not all S/S processes are conducive to the stabilization of all types of wastes.

Inorganic matrices are in most cases easier to stabilize than organics. Generally, organics do not react to become chemically part of the solid matrix, but remain entrapped in pores. Inorganics may either be entrapped or incorporated into the chemical structure, depending upon the treatment process.

Solidification/stabilization processes are generally used for treatment of contaminated sludges or soils. Major producers of hazardous sludge include private industries, utility companies, and water/wastewater treatment plants.

Table 3 is a compilation of a number of currently available S/S processes and applicable waste types. This table is based upon information identified in the literature search and provided by the S/S vendors responding to our survey.

FIXATION CHEMISTRY

Two of the principal types of S/S processes used for the fixation of metal-bearing wastes are discussed further to illustrate the principal mechanisms of stabilization, i.e., physical encapsulation and chemical fixation. One of these processes uses soluble silicates as the principal S/S ingredient. The other uses sodium sulfide and is sometimes referred to as the "Conner Process", after its inventor Jesse Conner. Vendors may use somewhat different versions of these processes. However, it is likely that

the basic process, which consists of S/S ingredient plus cement plus water, is similar. Process parameters, such as setting time, relative proportions of ingredients, sequence of addition, and pH may also be varied depending on the waste type. The mechanisms of fixation by silicate-based as opposed to sulfide-based processes are quite different and are summarized briefly below.

Silicate Process

As indicated above, the principal S/S ingredient is a "soluble silicate", almost always sodium silicate, which is about 5 times less expensive than the potassium salt. Sodium silicates are manufactured by fusing Na_2CO_3 and silica sand at 1,100 - 1,200 °C. The resulting product is an amorphous glass that can be dissolved under high pressures to produce hydrated sodium silicate in a variety of forms. In S/S applications, the sodium silicate is added as a concentrated solution, which has the appearance of a translucent syrup. Soluble silicates have numerous other industrial applications, and their role in waste stabilization is relatively new. They have also been used as binders, adhesives, surface active agents, detergency aids, and corrosion inhibitors (The PQ Corp., undated). Different products and grades have been developed for the various applications and are characterized by several basic properties, principally SiO_2/Na_2O weight ratio, percent solids, viscosity, and density. For example, the PQ Corporation manufactures a product referred to as "N"®, for S/S applications (The PQ Corp., 1988). "N"® is a syrupy liquid having a SiO_2/Na_2O ratio of 3.22, Na_2O content of 8.9 percent, SiO_2 content of 28.7 percent, density (at 20°C) of 1.38 g/cm^3, pH of 11.3, and viscosity of 180 centipoises (The PQ Corp., 1988).

The sodium silicate reagent is actually a mixture of solution species, including monomers (anionic forms of silicic acid), dimers, trimers, and larger multimeric species or polymers. Figure 1 illustrates the solubility and speciation of silica in water at pH 5 - 13. Polymeric species and colloids predominate above and to the left of the grey shaded
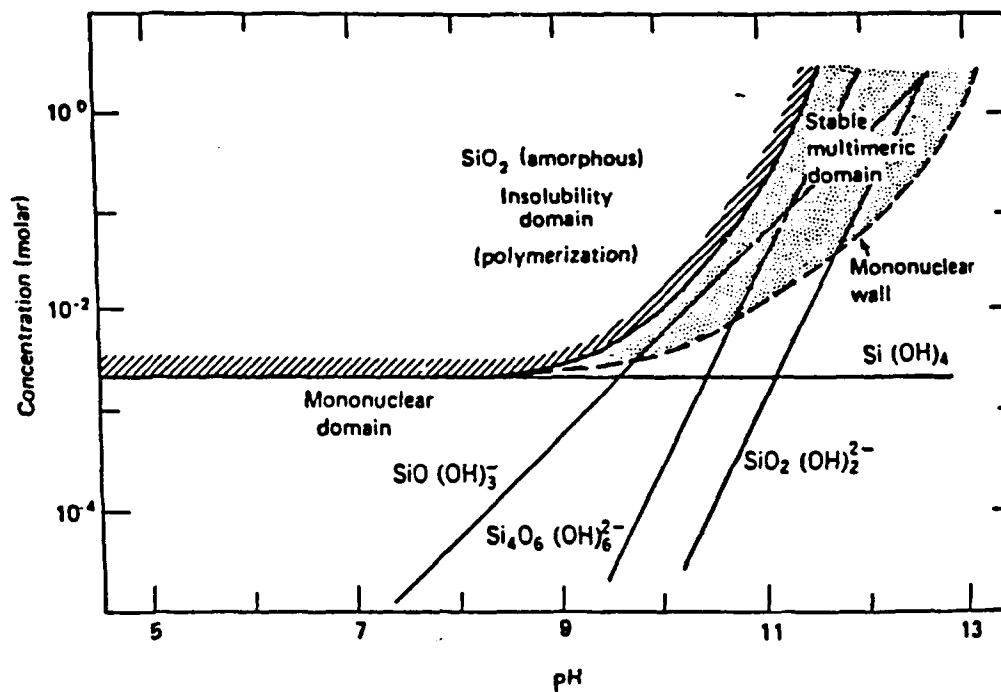
FIGURE 1. SOLUBILITY AND SPECIES IN EQUILIBRIUM WITH AMORPHOUS
SILICA (from Stumm and Morgan, 1970, p. 396)

region, corresponding to high silica concentrations and lower pH. Monomeric species, such as $Si(OH)_4$, $SiO(OH_3)^-$, and $SiO_2(OH_2)^{2-}$, predominate below and to the right of the grey shaded region, corresponding to either lower silica concentrations at lower pH or higher silica concentrations at higher pH. The third domain is indicated by the grey shaded region, where silica polyanions are stable. Commercially available concentrated silicate solutions plot in this multimeric domain (Stumm and Morgan, 1981). These solutions are supersaturated with respect to amorphous silica, and precipitation will eventually occur. The precipitation process will occur instantaneously upon even a slight lowering of the pH, which would cause the solution to move out of the shaded area in Figure 1 to the left into the insolubility domain.

This is the basis for the mechanism of stabilization by soluble silicate. When solutions of relatively high concentrations of soluble silicate are acidified, the silicate anions crosslink, polymerize, and form a gel. This gel coats and physically encapsulates the waste and forms a protective coating. Therefore, the primary mechanism of stabilization is physical encapsulation, and the process should be about equally effective for both metals and organics. In reality there is usually some degree of hydrolysis of metals that occurs because of the alkaline pH environment (typically pH 11 - 12). Consequently, the stabilization mechanism frequently includes chemical precipitation for certain metallic constituents that form insoluble precipitates upon hydrolysis. Thus, the degree of stabilization of certain metals is likely better than that for organics, which do not chemically react with the matrix.

The soluble silicate process results in a product which has more structural integrity than the original waste but which is not hardened like bricks. Typically, the silicate and cement are added in concentrations sufficient to create a blocky texture but not complete solidification. The product is easily disaggregated upon impact. However, encapsulation occurs on a microscopic rather than a macroscopic level. Therefore, good mixing during the stabilization process is important.

Sulfide Process

As indicated above, most versions of the so-called "Conner Process" utilize sodium sulfide (Na_2S) in combination with water and cement. The sodium sulfide may be added either as a solid or a solution. If it is added as a solid, then care must be taken to ensure adequate dissolution of the sodium sulfide so that the sulfide may react with the waste. There is also an EPA guideline that free sulfide concentrations (as determined by the so-called "Claussen test") in the waste should not exceed 500 ppm. Therefore, large quantities of undissolved sodium sulfide are also undesirable from this perspective.

If the sulfide is added as a solution, the total amount of sulfide that can be delivered to the waste is limited by the solubility of sodium sulfide in water, which is approximately 16.2 g Na_2S/100 g saturated solution at 22°C (Seidell, 1919). The dissolution of sodium sulfide in water is slightly exothermic.

Sodium sulfide is relatively alkaline and, when dissolved in pure water, will result in a solution having a pH of approximately 13.5. As shown in Figure 2, the pH at which the predominance of HS^- gives way to the S^{2-} species is 12.92 (Butler, 1964). Since sodium sulfide contains sulfide in the form of S^{2-}, the pH of pure sodium sulfide solutions will fall in the range of the S^{2-} predominance region in Figure 2.

The primary mechanism of fixation is the formation of sulfide salts. Numerous metals form very insoluble salts with the sulfide ion. The dissolution of metal salts is usually described by a solubility product, defined as follows. For the reaction:

$$A_mB_n(s) \rightarrow mA^{n+}(aq) + nB^{m-}(aq) \qquad (1)$$
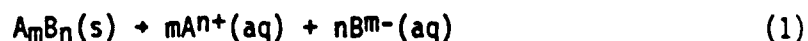
where A is the metal and B is the anion (in this case the S^{2-} ion), the conventional solubility product expression is:

$$K_{sp} = \left[A^{n+}(aq) \right]^m \left[B^{m-}(aq) \right]^n \qquad (2)$$
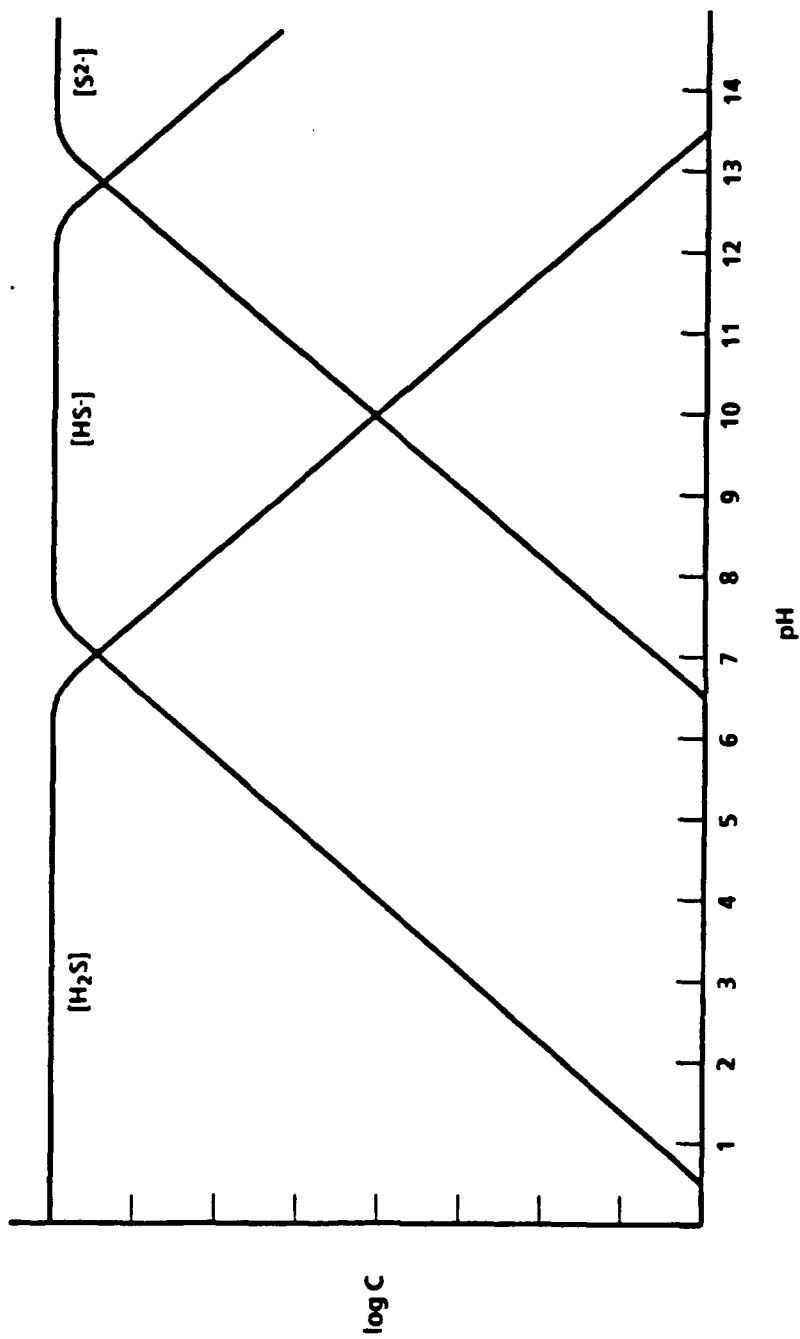
12

FIGURE 2.   SPECIATION DIAGRAM FOR SULFIDE SPECIES vs. pH

Therefore, the higher the solubility product K_{sp}, the more soluble the salt. Solubility products are typically expressed as their negative base-10 logarithms, pK_{sp}, where high values of pK_{sp} indicate low solubility (pK_{sp} values in excess of 20 indicate very low solubility). pK_{sp} values for selected metal sulfides are shown below (Butler, 1964):

Sulfide	pK_{sp}
Ag_2S	49.6
Cu_2S	48.0
Tl_2S	20.3
HgS (black)	51.8
HgS (red)	52.4
CuS	35.1
PbS	26.6
CdS	26.1
SnS	25.0
ZnS (sphalerite)	23.8
ZnS	21.6
CoS	22.1
NiS	20.7
FeS	17.3
MnS (green)	12.6
MnS (pink)	9.6

The formation of metal sulfides generally proceeds quite rapidly and, at elevated pH, the predominant sulfide species is S^{2-}, thus promoting the precipitation reaction. Depending upon the sulfide concentration and the acidity and buffering capacity of the waste, the pH of the treated waste will typically exceed 13, and it may be necessary to neutralize the waste prior to disposal in order to meet the RCRA corrosivity guidelines (maximum) of pH 12.5. This is usually accomplished after fixation rather than during fixation, to avoid inhibiting the fixation reaction by converting S^{2-} to HS^{-}. The additive is usually an inexpensive, weakly acidic salt with pH buffering capacity in the near neutral range. Sodium

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bicarbonate is frequently used. However, note from Figure 2 that excessive acidification (to pH <7) can lead to the liberation of H_2S gas if significant free sulfide (that is, not bound by metal) is present in the waste. The stability of the metal sulfides, however, should not be affected greatly by acidification to this pH range, because of the very insoluble nature of the metal sulfide precipitates (Barnes, 1979).

In that the primary mechanism of waste stabilization in the sulfide process is chemical (i.e., precipitation), and certain metals have a greater affinity for reacting with sulfide than others, it follows that the sulfide process is better suited for some wastes than others. A list of metals that form insoluble salts with sulfide was provided above. Others, for example chromium, selenium, and arsenic do not, although certain metals that do not precipitate with sulfide may hydrolyze at the elevated pH conditions or be reduced to a lower, less soluble valence state by the sulfide. Also, it stands to reason that the process would be relatively ineffective in stabilizing organics, though there have been vendor claims to the contrary.

Finally, the physical form of the waste changes relatively little during stabilization by sulfides. If the waste contains much iron, then the color will darken significantly because of the formation of iron sulfides. The cement adds to the structural integrity of the waste, but it disintegrates under relatively little force. A waste treated with sulfide will be noticeably less consolidated than the same waste treated with soluble silicate.

STABILITY OF THE TREATED WASTES

Stability of S/S waste is dependent upon: 1) physical isolation of the contaminant in a strong, durable structure; and/or 2) chemical immobilization of the contaminant in order to prevent leaching of the contaminant into the environment. This section is divided into four subsections corresponding to: 1) physical stability tests; 2) leaching methodologies; 3) factors affecting waste stability; and 4) stability data.

Physical Stability Tests

The primary focus of this review is on chemical stability. However, physical stability is briefly mentioned because S/S wastes disposed in a landfill may have to adhere to any applicable compressive strength guidelines. Standard concrete testing procedures are often used to test the physical stability of treated wastes. Table 4 lists commonly used procedures.

Leaching Methodologies

Two tests, the Extraction Procedure Toxicity (EP Tox) Test and the Multiple Extraction Procedure (MEP), are frequently used to evaluate the chemical stability of treated wastes. The tests are based on the leaching potential of a contaminant from the treated waste. The EP Tox test is a once-through batch leaching methodology which uses a weak acetic acid solution as the leachate. It is employed for evaluating shorter-term waste stability upon contact with water in the environment. The MEP test simulates the leaching that a waste in an improperly designed sanitary landfill will undergo from repetitive influxes of acid rain. Leachate in this case is a dilute sulfuric/nitric acid solution. The method is frequently used to infer the long-term stability of a waste in the environment in contact with groundwater. Each test is described in detail in EPA's SW-846 Solid Waste document. A new procedure, the Toxicity Characteristic Leaching Procedure (TCLP), has been developed and is similar to the EP Tox test, except it allows for the extraction of volatile organic compounds, using a zero-headspace extractor. The TCLP is conducted at a lower pH and is described as being more characteristic of a typical leaching situation.

Certain states have developed their own leaching methodologies. For example, the State of California recognizes the Waste Extraction Test (WET), which is a once-through batch leaching methodology similar to the EP

Tox test. Sodium citrate replaces acetic acid in the leachate. Both the WET test and the EP Tox test are used in the determination of whether a waste should be classified as hazardous.

Factors Affecting Waste Stability

Metal leaching from stabilized waste is controlled by many complex factors, including the type and speciation of the metal, treatment chemicals used, particle size in the stabilized waste, acid flux through the waste, and the time of contact with the leachate. Repeated contact with groundwater will inevitably lead to changes in waste chemistry, and some of these changes may affect the stability of the wastes.

From a geochemical perspective, sulfide-treated metal-bearing wastes should remain stable in a landfill unless 2 conditions exist simultaneously: (a) low pH; and (b) high Eh. Most metal sulfides are insoluble at either high pH coupled with low Eh, or high or low pH coupled with low Eh (Barnes, 1979). Low pH coupled with high Eh is undesirable because low pH favors the formation of ionic metal species and high Eh promotes oxidation of sulfide to sulfate. Landfill leachates frequently contain elevated concentrations of organic acids, and pHs as low as 3 - 4 are not unusual. However, the organic acids are by-products of anaerobic decomposition reactions, indicating the existence of low Eh conditions. Hence, it seems unlikely that sulfide waste, as long as it is properly buried in the landfill, would encounter high Eh. However, it may be inadvisable to use sulfide waste as daily cover, where the combination of low pH and high Eh conditions might be encountered.

Certain impurities can affect the strength, durability, and permeability of Portland cement and asphalt mixtures. Organics containing hydroxyl or carboxylic functional groups, for instance, may delay or completely inhibit the pozzolanic or Portland cement-based reactions responsible for solidification (Wiles, 1987). Temperature, humidity, and mixing also affect the stability of the treated waste during setting of cement-based processes. Extremely cold environments inhibit cementitious

reactions. Temperatures above 66°C may destroy the reactions that result in setting (Wiles, 1987). High humidity may accelerate setting. Extensive mixing may destroy the initial set of the stabilized material and result in a low strength product (Wiles, 1987). Thus, the precise reproducibility of a given S/S process is low because of the numerous factors involved.

Stability Data

Methods of reporting leaching data lack consistency; therefore, it is very difficult to compare data sets from one vendor (or author) with that of another. Table 5 summarizes qualitative statements made by the various vendors/authors on the stability of their treated wastes.

Data on long-term stability are limited. The majority of data on stability has been obtained using a once-through leaching methodology (such as the EP Tox test) which is not intended to address the potential for long-term leaching of a waste (Bishop, 1988). MEP testing, on the other hand, does assess longer-term stability; however, few data are currently available. The MEP test flushes the waste with large volumes of water, but does not simulate chemical reactions having slow kinetics that might take place over a period of years or decades or more. Computer modeling may also provide a means of predicting long-term stability.

One available site-specific study on long-term stability was provided by Landreth (1981), who examined soils and groundwaters underlying stabilized waste in four different disposal sites around the United States approximately seven to eight years after disposal. The original wastes, which consisted of metal finishing, electroplating, and refinery sludges, contained a wide variety of heavy metal pollutants and were stabilized using a proprietary lime-silicate process. Changes in groundwater quality indicators (e.g., sodium, chloride, sulfate, boron, cyanide, and others) related to the waste disposal activities could be observed at 3 of the 4 sites investigated. Metal contamination in groundwater and soils underlying the waste could not be detected at levels that presented a serious pollution problem. However, distilled water extracts of these soils

showed that, while metal concentrations were low, the proportion of soluble metal to total metal was surprisingly high, leading to the suggestion that some of the heavy metals were escaping from the waste and being held in the soils in a readily leachable state.

POTENTIAL USE/REUSE OF TREATED WASTE

Treated hazardous waste is generally disposed of in a landfill. Stricter landfill regulations have stimulated interest in alternatives to landfill disposal. Certain wastes can be solidified and used as construction or road-building materials. Benson et al. (1985), for instance, solidified sand blasting residue (containing elevated levels of cadmium and lead) with concrete to produce a treated waste that had suitable design strength and enough chemical stability to be used in construction. Certain wastes are suitable for use as filler in asphalt. In this case the product is directly reusable as paving material. Table 6 provides information on S/S products that are reported to have potential reuse value.

Alternate reuses including landfill disposal are likely to be subject to approval from cognizant regulatory agencies. There is likely some risk involved in reuses other than disposal, because of the possible liability that could result from the lack of long-term stability. Also, if the stabilized waste does become destabilized at some point in the future, then it may be reclassified as hazardous waste, hence negating the effects of the initial treatment.

WASTE STABILIZATION COSTS

Waste stabilization costs are dependent upon the type, quantity, and complexity of the waste. Labor, transportation, and equipment costs must be evaluated in addition to the cost of the reagents and additives used in the process. Table 7 summarizes the cost information that was obtained from the vendor survey and the literature search. These figure are not

directly comparable as they have not been normalized to the same set of cost elements. They do, however, give an indication of the average costs charged by many of the current vendors.

RELEVANT DoD ACTIVITIES

Solidification and stabilization of hazardous wastes have been investigated by both the private sector and public agencies. The Army has conducted at least three major S/S studies in recent years. The studies were conducted at the Environmental Laboratory of the U.S. Army Engineer Waterways Experimental Station in Vicksburg, Mississippi. The first S/S study provided: 1) a discussion of various S/S processes; 2) a summary of stabilized waste properties; 3) guidelines for evaluating stabilized waste; and 4) a vendor list (U.S. Army Engineer Waterways Experimental Station Environmental Laboratory, 1980). The second study contained: 1) information on various S/S methods; 2) a summary of the criteria for process selection; 3) a discussion of environmental concerns; and 4) procedures for the closure/clean-up of a hazardous waste site (Cullinane et al., 1986). The Army's most recent work has focused on identifying constituents that interfere with various S/S processes (Jones, 1988).

The Air Force was also contacted for information regarding relevant work in this area. Most of their work has focused on treatment processes such as biodegradation rather than solidification (Matuszko, 1989).

TABLE 1. TYPICAL CHEMICAL COMPOUNDS IN POZZOLANS AND PORTLAND CEMENT

Chemical Compound	Pozzolan (%)	Cement (%)
SiO_2	39.9 - 58.2	22.6
Al_2O_3	16.7 - 25.8	4.3
Fe_2O_3	5.8 - 9.3	2.4
CaO	3.3 - 24.3	64.4
MgO	1.8 - 4.6	2.1
SO_3	0.6 - 3.3	2.3
Na_2O and K_2O	0.6 - 1.3	0.6

Source: Pozzolanic Technical Bulletin No. 7 as referred to in vendor information from Solidtech, Inc.

TABLE 2. COMPATIBILITY OF SELECTED WASTE TYPES WITH WASTE SOLIDIFICATION/STABILIZATION TECHNIQUES

Waste Component	S/S Treatment Type			
	Cement-based	Lime-based	Thermoplastic Solidification	Organic Polymer (UF)*
Inorganics				
1. Heavy metals	Compatible	Compatible	Compatible	Acid pH solubilizes metal hydroxides
2. Oxidizers	Compatible	Compatible	May cause matrix breakdown, fire	May cause matrix breakdown
3. Sulfates	May retard setting and cause spalling unless special cement is used	Compatible	May dehydrate and rehydrate causing splitting	Compatible
4. Halides	Easily leached from cement, may retard setting	May retard set, most are easily leached	May dehydrate	Compatible
5. Acid wastes	Cement will neutralize acids	Compatible before incorporation	Can be neutralized	Compatible
6. Radioactive materials	Compatible	Compatible	Compatible	Compatible
Organics				
1. Organic solvents and oils	Many impede setting, may escape as vapor	Many impede setting, may escape as vapor	Organics may vaporize on heating	May retard set of polymers
2. Solid organics (e.g., plastics, resin, tars)	Good--often increases durability	Good--often increases durability	Possible use as binding agent	May retard set of polymers

*Urea-Formaldehyde resin.

Source: SW-872, U.S. EPA, 1982 (complete ref on p. 5 of the Wiles & Apel report).

TABLE 3. SUMMARY OF STABILIZATION PROCESSES AND APPLICABLE WASTE TYPES

Hazardous Constituent/Waste Type	Stabilization Process/Formulation	Reference/Vendor
Heavy metals in sandblast waste	pozzolanic process (Tradename additives used include Calicox and Thiosorbic)	Dravo Lime Company, 1988
Heavy metals and organics in soil	grout slurry produced by cement-based process in which metals are stabilized as insoluble metal silicates	FPL/Qualtec, Inc., 1988
Heavy metals and organics in acidic sludge	pozzolanic F/E/S formulation	Enreco, Inc., 1988
Heavy metals and organics in soil/ sludge	pozzolanic process which produces a slurry	Soliditech, Inc. 1988
Heavy metal and organics in soft soils	a neutral/nontoxic form of silica is used in combination with Portland cement	Trident Engineering Associates, Inc., 1988
Heavy metals and organics in soil/sludge	inert artificial clays are produced through chemical fixation/solidification	McLaughlin Enterprises, Inc., 1988
Heavy metals and organics in soil/liquid sludge	a system (containing soluble silicates and silicate setting agents) that produces a non-polluting gel which solidifies to form a clay-like solid	Chemfix Technologies, Inc., 1988
Heavy metals and organics in soil/sludge	a silicate-based system used in combination with lime, Portland cement, fly ash and/or kiln dust	Lopat Enterprises, Inc. 1986

TABLE 3. SUMMARY OF STABILIZATION PROCESSES AND APPLICABLE WASTE TYPES (Continued)

Hazardous Constituent/Waste Type	Stabilization Process/Formulation	Reference/Vendor
Inorganic metals and organics	a solidification process (developed by Waste Chem Corporation) which uses asphalt binders and extrusions	Wiles and Howard, undated
Inorganics and organics in sludge	a cement-based process referred to as the "chloranan 20" solidification process developed by Hazcon Inc.	Hill, 1987
Inorganics and organics in sludge	a chemical fixation process which involves bonding of hazardous materials with sulfo-ferri-aluminate hydrates	Hill, 1987
Inorganics and organics in sludge/soil	a solidification/stabilization process which utilizes a surface modified alumina silicate sorbent followed by a silicate solidifying agent	Silicate Technology Corporation, 1988
Inorganics and organics in soil	in situ fixation and stabilization by injecting and mixing of various reagents and additives into the contaminated soil	Toxic Treatment (USA) Inc., 1988
Inorganics and organics	a silicate-based process (developed by Stablex Corp)	Telles et al, 1984
Inorganic and organic wastes in sludge	a pozzolanic process (developed by I.O. Conversion Systems)	Telles et al, 1984

TABLE 3. SUMMARY OF STABILIZATION PROCESSES AND APPLICABLE WASTE TYPES (Continued)

Hazardous Constituent/Waste Type	Stabilization Process/Formulation	Reference/Vendor
Inorganic and organics	a process (developed by International Waste Technologies) which utilizes a silicate-colloid or an inorganic polymer that chemically bonds organics and treats inorganics	Wiles C. C. and H. K. Howard
Inorganics and organics	a process (developed by Tennessee Valley Authority) which uses fluidized bed combustion with finely divided silica	Wiles, undated
Inorganics and organics	a process which converts hazardous waste to a granular solid	Chem-Met Services, Services, 1989
Radionuclides, metals and organics in soils, sludges and ponds	a solidification process (developed by American Coal Ash Association, Inc.) which uses lime and coal fly ash	Wiles, undated
Radioactively contaminated soil	in situ vitrification	Jacobs, undated
Low-level radioactive sludge and other wastes	Radlok high integrity containers constructed of high density, cross-linked polyethylene	Hittman Nuclear Systems, 1988
Low-level radioactive waste contained in bead and powder resin, diatomaceous earth, filter sludge, boric acid and sodium sulfate concentrate, oil, and/or abrasive decontamination grit	an in-container, cement-based solidification system	Hittman Nuclear Incorporated, 1988

TABLE 3. SUMMARY OF STABILIZATION PROCESSES AND APPLICABLE WASTE TYPES (Continued)

Hazardous Constituent/Waste Type	Stabilization Process/Formulation	Reference/Vendor
Low-level radioactive soil	in situ chemical grouting containing either sodium silicate or acrylamide used in combination with a high density polyethylene barrier	Cox, undated
Material dredged from lakes and rivers	a process (developed by Continental Dredge and Marine Corporation) in which a barge-mounted incinerator combusts dredged material; the incinerator ash is then solidified	Wiles, undated
Organic waste and ion exchange resins	a process (developed by Penberthy Electro-mett International) which fixes waste in molten glass	Telles et al, 1984
Ion exchange resins	a process (developed by Suntrac Services) which fixes waste by using a proprietary long-chain polymer	Telles et al, 1984
Oil and organic wastes	a process (developed by Suntrac Services) which fixes waste using proprietary polymers and Portland cement	Telles et al, 1984
Oil and organic wastes; ion-exchange resin bed	a gypsum/polymer process (developed by U.S. Gypsum Co.)	Telles et al., 1984
Ion-exchange beads and filter media	cement-based process (developed by Atcor Washington Inc.)	Telles et al., 1984
Waste oil	a process (developed by Delaware Custom Material Inc.) that uses cement, shale silicate, and proprietary emulsifiers	Telles et al, 1984

TABLE 3. SUMMARY OF STABILIZATION PROCESSES AND APPLICABLE WASTE TYPES (Continued)

Hazardous Constituent/Waste Type	Stabilization Process/Formulation	Reference/Vendor
PCB laden oil	a lime-based process (developed by Sludge Master)	Telles et al, 1984
Utility flue gas desulfurization sludge	pozzolanic process (Tradename additives used include Calicox and Thiosorbic)	Dravo Lime Company, 1988
Cladding-removal waste	cement-based grout	VFL Technology Corporation 1988
Sludge and ash from wastewater treatment	lime-based technique compared to cement-based technique	Malone et al, 1985
Acidic hydrocarbon sludge and spent clay	pozzolanic process	Martin, undated

TABLE 4. STANDARD TEST METHODS FOR PHYSICAL PROPERTIES

Test	Source
Bulk and dry unit weight	Appendix II of EM 1110-2-1906*
Unconfined compressive strength	Appendix XI of EM 1110-2-1906 and ASTM Method D2166-66**
Permeability	Appendix VII of EM 1110-2-1906
Wet/dry durability	ASTM Method D559-57
Freeze/thaw durability	ASTM Method D560-57

Reference: U.S. Army Engineer Waterways Experimental Station Environmental Laboratory, 1980.

TABLE 5. WASTE STABILITY DATA

Reference/Vendor	Process	Leaching Comments
ToxCo Inc., (formerly McLaughlin Enterprises) 1988	system based on use of sulfides, soluble silicates and silicate setting agents	no reversal of the treated materials at sites that have been treated 6 to 7 years earlier
Telles et al, 1984	sodium silicate & Portland cement system investigated by Telles & Lubowitz	meets EPA criteria with loading of up to 24,000 ppm of Pb, Cd, Cr
Landreth et al, 1981	cement-based process used to solidify and stabilize metal finishing waste, electroplating waste, and refinery sludge	distilled water extracts of sub-waste soils had elevated levels of Cr, Pb, & Se suggesting that contaminants from waste could be leaching into groundwater
Chemfix Technologies, Inc., 1988	system based on soluble silicates and silicate setting agents	metals converted to stable non-leachable metal silicates; however, anionic and nonionic metals are not retained by Chemfix treated materials
FPL/Qualtec, Inc., 1988	Portland cement-fly ash system	metals stabilized as metal silicates; grout can be poured or pumped into standing water without releasing contaminants
Trident Engineering Associates Inc., 1988	silica used in combination with Portland Cement	reduces leaching of heavy metals and other hazardous materials to a generally acceptable level
Benson et al, 1985	concrete-based technique	cadmium contained in waste is effectively fixed in the solid concrete matrix

TABLE 5. WASTE STABILITY DATA (Continued)

Reference/Vendor	Process	Leaching Comments
Hazcon, Inc., 1989	a cement-based process which utilizes chloranan (a polymer-based chemical)	priority pollutant metals ranging from 3000 ppm to 24,000 ppm were successfully immobilized...migration reduction potentials for leachates exceed 98%
Silicate Technology Corporation, 1988	technique uses a surface-modified alumina-silicate sorbent followed by a silicate solidifying agent	data include TCLP results for benzene, toluene, and xylene and EP Tox results for Pb and Cd
Lopat Enterprises, Inc., 1988	cementitious fixative	90-99% reduction in hazardous material in leachate
Telles et al, 1984	calcium-polysilicate system investigated by Telles & Lubowitz	acceptable leach performance was only obtained at or below 0.2% loading
Telles et al, 1984	patented silicate-based technology developed by Stabley Corporation	<1 ppm of toxicants lost to water in leaching tests
Telles et al, 1984	encapsulation technique involving polybutadiene and polyethylene to treat an organic pesticide	no contaminants in leachate solution
Sanning et al, 1985	asphaltic system for treating dioxin-contaminated soils	low water solubilities of dioxins will limit leaching into environment
Telles et al, 1984	vinyl esters were used to treat ion exchange material in a study by Smitton and Haigton	vinyl ester leach rates were an order of magnitude lower in some instances than polyester/cement-bound wastes

TABLE 5. WASTE STABILITY DATA (Continued)

Reference/Vendor	Process	Leaching Comments
Telles et al, 1984	urea-formaldehyde resins used to treat low-level radioactive waste such as beads and powder resins	free liquid possible stability problem
Telles et al, 1984	a cement-based radwaste system developed by Atcor Washington	acceptable leach rates for shallow-land burial
Hittman Nuclear, 1988	cement system	meets all requirements for leach resistance

TABLE 6. SELECTED REUSES FOR SOLIDIFICATION/STABILIZATON WASTES

Process	Waste Type	Vendor/Reference	Reuse Potential
Cement-based process	Sandblast residue containing heavy metals	Benson et al., 1985	Sandblast residue is used as a concrete additive in producing common construction materials
Pozzolanic process	Acidic sludge containing heavy metals and organics	Enreco, Inc., 1988	Final S/S material is used as landfill
S/S process (which utilizes soluble silicates and silicate setting agents) that produces a nonpolluting gel which solidifies to form clay-like solids	Soil or sludge containing heavy metals, polynuclear aromatic hydrocarbons, oil and grease, semivolatile organics, cyanide, and/or chlorinated pesticides	Chemfix Technologies Inc., 1988	Clay-like solids can be used as: 1) an agricultural lime substitute, 2) sanitary landfill, or 3) structural landfill
Chemical fixation/solidification is used to produce inert artificial "clays"	Municipal sewage, refinery wastes, drilling muds, chemical plant residues, and heavy metals	ToxCo Inc., 1988	Artificial "clay" is reportedly suitable for landfill or agriculture
S/S process which utilizes a silicate sorbent and a silicate solidifying agent	Soil or sludge contaminated with organics and/or inorganics	Silicate Technology Corporation, 1988	Final S/S material is used as landfill

TABLE 7. WASTE SOLIDIFICATION/STABILIZATION PROCESS COST COMPARISON

Process	Cost	Reference/Vendor
Pozzolanic F/E/S formulation	$9-35/yd³	Enreco, Inc., 1988
Pozzolanic process which produces a slurry	$70/ton* (includes labor, mixing, and equipment cost)	Soliditech, Inc., 1988
Pozzolanic process	$60/ton* (plus transportation)	Dravo Lime Company, 1988
Pozzolanic process used by I.U. Conversion Systems	$10-40/ton	Telles, et al., 1984
Pozzolanic process	$60/ton	Bell et al., 1981
Cement-based process	$60/ton	Bell et al., 1981
In situ vitrification	$400/m³ (19?? dollars)	Brouns, R. A. and C. L. Timerman, 1982 as cited in Telles et al, 1984
In-container, cement-based solidification system	$75/ft3*	Hittman Nuclear Systems, 1988
In situ chemical grouting (containing either sodium silicate or acrylamide) used in combination with a high-density polyethylene barrier	$13/ft3	Cox, undated
Proprietary cement-based processes	$70/yd3 (approx)	ToxCo Inc., undated

TABLE 7. WASTE SOLIDIFICATION/STABILIZATION PROCESS COST COMPARISON (Continued)

Process	Cost	Reference/Vendor
Cement-based grout	$20-$50/ton*	VFL Technology Corporation, 1988
Cement-based grout slurry in which metals are stabilized as insoluble metal silicates	$80-$150/ton*	FPL/Qualtec, Inc., 1988
Portland cement is used in combination with a neutral/nontoxic form of silica	$1.50-$3.00/yd3 (plus labor, handling, mixing & machinery costs)	Trident Engineering Associates, 1988
A patented silicate-based system developed by Lopat Enterprises, Inc. and used in combination with lime, Portland cement, fly ash and/or kiln dust	$20-$200/ton	Mineral By-Products Inc., 1988
A system (containing soluble silicates and silicate setting agents) that produces a non-polluting gel which solidifies to form a clay-like solid	$30-$50/yd3	Chemfix Technologies, Inc., 1988
A solidification/stabilization process which utilizes a surface modified alumina-silicate sorbent followed by a silicate solidifying agent	$30-$50/ton	Silicate Technology Corporation, 1988
A silicate-based process developed by Stablex Corp.	$5-$350/ton (dependent upon type, quantity and complexity of waste)	Telles et al., 1984

TABLE 7. WASTE SOLIDIFICATION/STABILIZATION PROCESS COST COMPARISON (Continued)

Process	Cost	Reference/Vendor
Thermoplastic solidification (dried waste is mixed with a heated plastic matrix such as bitumen)	$392/ton	Bell et al., 1981
Organic polymer processes (waste is mixed with an organic resin such as urea-formaldehyde and then polymerized with a catylst)	$554/ton	Bell et al., 1981
Surface encapsulation using high-density polyethylene	$90/ton	Bell et al., 1981

* Verbal quotes from vendor representatives.

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APPENDIX A

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